Space-time Phase Transitions in Driven Kinetically Constrained Lattice Models

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Kinetically constrained models (KCMs) have been used to study and understand the origin of glassy dynamics. Despite having trivial thermodynamic properties, their dynamics slows down dramatically at low temperatures while displaying dynamical heterogeneity as seen in glass forming supercooled liquids. This dynamics has its origin in an ergodic-nonergodic first-order phase transition between phases of distinct dynamical "activity". This is a "space-time" transition as it corresponds to a singular change in ensembles of trajectories of the dynamics rather than ensembles of configurations. Here we extend these ideas to driven glassy systems by considering KCMs driven into non-equilibrium steady states through non-conservative forces. By classifying trajectories through their entropy production we prove that driven KCMs also display an analogous first-order space-time transition between dynamical phases of finite and vanishing entropy production. We also discuss how trajectories with rare values of entropy production can be realized as typical trajectories of a mapped system with modified forces.

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Introduction. – The transformation of a supercooled liquid into an amorphous solid glass at low temperatures has fascinated scientists for decades [1], yet a comprehensive theory explaining the microscopic origins of this "glass transition" is still missing. Several scenarios are advocated in the literature. These include: (i) A transition to an "ideal" glass state reached at some critical value of a thermodynamic parameter such as the temperature. This transition may be thermodynamic, as in the random first-order transition theory [2], or kinetic, as in mode-coupling theory [3]. (ii) An "avoided" glass transition, controlled by an unreachable thermodynamic critical point [4]. (iii) The glass transition as a purely dynamical phenomenon that is not controlled by structural changes and not related to a phase transition in a thermodynamic sense. This approach [5] considers dynamic heterogeneity [6], the observation of large spatial and temporal fluctuations in "dynamical activity", to be the crucial feature of glass formers and derives most of its predictions and insights from the study of kinetically constrained models (KCMs) of glasses [7].

Analytical studies of KCMs [8] and numerical investigations of atomistic glass forming fluids [9] have shown the existence of a true phase transition in ensembles of trajectories. This "space-time" phase-transition is a first-order transition between two dynamical phases which differ in their overall "dynamical activity" [8, 9]. This transition is not controlled by thermodynamic fields. To reveal it one needs to employ a statistical mechanics of trajectories whereby distributions of trajectories are studied by controlling either dynamical activity or a conjugate nonequilibrium field s [8–11]. Emergence of dynamic heterogeneity and other fluctuation phenomena can be seen as manifestations of this underlying transition [5].

In this Letter we show the existence of analogous space-

time phase transitions in glassy systems driven to nonequilibrium stationary states by non-conservative forces. In this case the phase-transition is between dynamical phases with finite and with vanishing entropy production. We do so by studying ensembles of trajectories via the large-deviation method in a class of KCMs driven away from equilibrium by non-conservative forces. Such forced KCMs [12, 13] can be motivated experimentally by, for example, pulling a tracer particle through a supercooled liquid or a colloidal suspension [14]. The largedeviation method allows to study sub-ensembles of trajectories with non-typical values of dynamical observables in a way that sheds light onto the phase structure of the dynamics. We also show that these sub-ensembles of rare trajectories can be generated as typical trajectories by a mapping to an alternative system with modified forces. Such a mapping might allow to bridge the conceptual gap between computer generated biased ensembles and the experimental observation of space-time phase transitions.

Biased ensembles of trajectories.— We first recall the results for stationary equilibrium dynamics [8]. The activity K [8–11] is a dynamical (space-time) order parameter. Information about the dynamics is encoded in the distribution of K, or equivalently, in the dynamical partition sum $Z(s) \equiv \langle e^{-sK} \rangle_0$. The brackets $\langle \cdots \rangle_0$ denote the average over trajectories in the original ensemble, i.e., it is a sum over trajectories C(t) weighted by $P_0[C(t)]$. The field s is conjugate to the activity and the factor e^{-sK} enhances the weight of trajectories with either higher than typical activity (s < 0), or with lower than typical activity (s > 0). One can think of s as giving rise to a biased ensemble of trajectories with weights $P_s[C(t)] \propto P_0[C(t)]e^{-sK[C(t)]}$. We refer to this biased ensemble as the s-ensemble [9].

Denoting N the system size and $t_{\rm obs}$ the length of trajectories, their product $Nt_{\rm obs}$ is the "volume" in spacetime. For large $Nt_{\rm obs}$, the dynamical partition sum acquires a large deviation form [15] with $Z(s; Nt_{\rm obs}) \sim$ $e^{Nt_{\text{obs}}\psi(s)}$. The large-deviation function $\psi(s)$ generates the moments of K in the biased ensemble, e.g., the mean activity rate is $\langle K \rangle / (Nt_{\rm obs}) = -\psi'(s)$ where $\langle \cdots \rangle$ now denotes the average in the s-ensemble. The function $\psi(s)$ is akin to a free-energy. For KCMs it has a first-order singularity at s=0 indicating a space-time phase transition [8]. This transition is between an ergodic phase of finite activity rate and a nonergodic phase of vanishing activity rate. In the nonergodic phase the system, through the kinetic constraints, is able to arrange trajectories such that the activity grows sublinearly. These trajectories "win" at $s = 0^+$ because active trajectories with nonzero activity acquire a negative weight in the partition sum and thus are exponentially suppressed for large $t_{\rm obs}$. At $s=0^-$ the opposite occurs and the dominant phase is the active, ergodic, one.

We now extend the s-ensemble approach to driven systems. Transitions between configurations \mathcal{C} occur with rates $w(\mathcal{C} \to \mathcal{C}')$ implying the exit rate $r(\mathcal{C}) \equiv \sum_{\mathcal{C}'} w(\mathcal{C} \to \mathcal{C}')$. For a given trajectory $\mathcal{C}(t)$ of time span t_{obs} the system undergoes K transitions $\mathcal{C}_{\alpha-1} \to \mathcal{C}_{\alpha}$ at times t_{α} where \mathcal{C}_0 is the initial state, i.e., $\mathcal{C}(t) \equiv (\mathcal{C}_0, \ldots, \mathcal{C}_K)$. The entropy produced in the environment in a single transition $\mathcal{C} \to \mathcal{C}'$ is $\Delta s(\mathcal{C}, \mathcal{C}') = \ln[w(\mathcal{C} \to \mathcal{C}')/w(\mathcal{C}' \to \mathcal{C})]$. In analogy to the activity, we may bias trajectories using the time-extensive medium entropy production $s_{\mathrm{m}}[\mathcal{C}(t)] \equiv \sum_{\alpha=1}^K \Delta s(\mathcal{C}_{\alpha-1}, \mathcal{C}_{\alpha})$, where we sum over all configuration changes. The corresponding dynamical partition function is

$$Z(\lambda; Nt_{\rm obs}) \equiv \langle e^{-\lambda s_{\rm m}} \rangle_0 \sim e^{Nt_{\rm obs}\Psi(\lambda)}$$
 (1)

where λ is the parameter conjugate to $s_{\rm m}$. In analogy with the activity, the mean entropy production rate in the λ -ensemble is given by $\langle s_{\rm m} \rangle / (N t_{\rm obs}) = -\Psi'(\lambda)$. The partition sum (1) has a natural transfer matrix representation, and $\Psi(\lambda)$ is given by the largest eigenvalue of the operator [16]

$$\mathbb{W}_{\lambda}(\mathcal{C}',\mathcal{C}) = \left[\frac{w(\mathcal{C}' \to \mathcal{C})}{w(\mathcal{C} \to \mathcal{C}')}\right]^{\lambda} w(\mathcal{C} \to \mathcal{C}') - r(\mathcal{C})\delta_{\mathcal{C}\mathcal{C}'}. \quad (2)$$

In what follows we study the dynamical phase structure of driven KCMs by calculating their large-deviation functions $\Psi(\lambda)$ from the corresponding operators \mathbb{W}_{λ} .

Driven Fredrickson-Andersen model.— The simplest KCM that displays glassy features is the one-spin facilitated Frederickson-Andersen (FA) model [7, 18]. We introduce a driven variant of the FA model with evolution operator

$$\mathbb{W}_0 \equiv \sum_{\langle ij \rangle} \left\{ \left[c a_i^{\dagger} + a_i - (c + \hat{n}_i) \right] \hat{n}_j + k_{i \to j} (a_j^{\dagger} a_i - \hat{n}_i) \right\}.$$

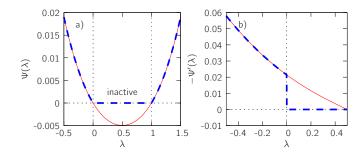


FIG. 1: (a) Mean-field estimate of the large-deviation function $\Psi^{(\mathrm{mf})}(\lambda)$ [Eq. (5), bold dashed] for the driven FA model with $c=0.2,\ k^-/k^+=0.7$. There is a first-order transition from an active (entropy producing) dynamical phase to an inactive dynamical phase at $\lambda=0$. The "reentrant" transition at $\lambda=1$ is due to the Gallavotti-Cohen symmetry. The thin solid line corresponds to the active branch for all values of λ . (b) Mean entropy production rate $-\Psi'(\lambda)$.

For simplicity we consider a bosonic version of the FA model, i.e. we allow for multiple occupancy of sites [17]. The sum runs over all pairs of nearest neighbors. Here, a_i^{\dagger} and a_i are creation and annihilation operators at site i with number operator $\hat{n}_i = a_i^{\dagger} a_i$. The first term describes the creation and annihilation of excitations at site i with rate $c \equiv e^{-\beta J}$, where J is the energy scale and $\beta \equiv 1/(k_{\rm B}T)$ is the inverse temperature. The dynamics is constrained in the sense that at least one nearest neighbor must be excited to allow transitions at site i. The second term allows for explicit diffusion of excitations from site i to one of its neighboring sites with rates $k_{i \to j}$ [19]. Using Eq. (2), the time-evolution operator for the λ -ensemble reads

$$W_{\lambda} = \sum_{\langle ij \rangle} \left\{ \left[\left(\frac{\hat{n}_{i}}{c} \right)^{\lambda} c a_{i}^{\dagger} + \left(\frac{c}{\hat{n}_{i}} \right)^{\lambda} a_{i} - (c + \hat{n}_{i}) \right] \hat{n}_{j} + k_{i \to j} \left[\left(\frac{k_{j \to i} \hat{n}_{j}}{k_{i \to j} \hat{n}_{i}} \right)^{\lambda} a_{j}^{\dagger} a_{i} - \hat{n}_{i} \right] \right\}. \quad (3)$$

We restrict our calculation to a one-dimensional lattice with N sites and periodic boundaries. Further, we consider spatially homogeneous rates: k^+ for a hop $i \to i+1$ and k^- for $i \to i-1$ such that the steady state solution for the mean density becomes independent of the site. The largest eigenvalue of (3) can be estimated in a mean-field approximation by maximizing the function $W(\bar{\phi},\phi)$ obtained from the (normally ordered) operator (3) by replacing $a^\dagger \to \bar{\phi}$ and $a \to \phi$. There are two solutions to the Euler-Lagrange equations $\partial W/\partial \phi = 0$ and $\partial W/\partial \bar{\phi} = 0$. In terms of the mean density $n = \bar{\phi}\phi$ the two solutions read: n = 0 and $n = (c/16)(3 + \sqrt{1 + 4\kappa(\lambda)/c})^2$. They correspond to the inactive and active phases, respectively. The λ -dependence is contained in

$$\kappa(\lambda) \equiv \left(\frac{k^+}{k^-}\right)^{\lambda} k^- + \left(\frac{k^-}{k^+}\right)^{\lambda} k^+ - (k^+ + k^-). \tag{4}$$

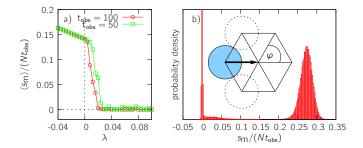


FIG. 2: (a) Numerical result for the mean entropy production rate in the (2)-TLG for parameters: L=16, $\rho=0.5$, and f=0.5. (b) Bimodal distribution of the entropy production rate at $\lambda=0.003$. The inset shows the kinetic constraint of the (2)-TLG: the particle can only move along the arrow if the two indicated sites are vacant.

The mean-field large deviation function becomes

$$\Psi^{(\mathrm{mf})}(\lambda) = \begin{cases}
0 & (0 \leqslant \lambda \leqslant 1) \\
2n^{3/2}(\sqrt{n} - \sqrt{c}) & (\lambda \leqslant 0 \text{ and } \lambda \geqslant 1).
\end{cases} (5)$$

The function $\Psi^{(\mathrm{mf})}(\lambda)$ and its first derivative are plotted in Fig. 1. The negative first derivative is the mean entropy production rate. It shows a discontinuous transitions at $\lambda=0$ between finite entropy production ($\lambda<0$, the active phase) and vanishing entropy production ($\lambda>0$, the inactive phase). For $\lambda=1$, there is a second "reentrant" transition to an active phase now with a negative entropy production, see Fig. 1(a). This second transition is a reflection of the transition at $\lambda=0$ due to the Gallavotti-Cohen symmetry, which enforces the relation $\Psi(1-\lambda)=\Psi(\lambda)$ on the large deviation function [16, 20] through Eq. (4). Biasing with $\lambda>1$ corresponds to driving that essentially changes the sign of all currents due to time-reversal symmetry.

Driven constrained lattice gas. – For a numerical study, we turn to a driven variant [12, 13] of a kinetically constrained triangular lattice gas (TLG) [7, 21]. Lattice gas particles diffuse on a two-dimensional triangular lattice with periodic boundary conditions. The density of particles ρ is conserved. A particle can only move to a neighboring site if the two adjacent sites are vacant, see inset of Fig. 2(b). This version of the model is called the (2)-TLG [21]. In addition, we apply a force f in the xdirection. As a consequence the rates for allowed moves are $w(\varphi) = e^{(f/2)\Delta x_{\varphi}}$ where $\varphi = 0, \pm \pi/3, \pm 2\pi/3, \pi$ is the angle between the displacement vector and the xaxis. In a single transition a particle moves in the xdirection the distance $\Delta x_{\varphi} = 2\cos\varphi = \pm 1, \pm 2$. We denote by $K_{\varphi}[\mathcal{C}(t)]$ the number of moves in the direction φ in a trajectory $\mathcal{C}(t)$. The activity is then given by $K[\mathcal{C}(t)] = \sum_{\varphi} K_{\varphi}[\mathcal{C}(t)],$ and the entropy production by $s_{\mathbf{m}}[\mathcal{C}(t)] = f \sum_{\varphi} K_{\varphi}[\mathcal{C}(t)] \Delta x_{\varphi}.$

We use transition path sampling [22] to numerically generate biased ensembles of trajectories, for details see Ref. [9]. Fig. 2(a) shows the mean entropy production

rate $\langle s_{\rm m} \rangle/(Nt_{\rm obs})$ per particle, from ensembles of 10^5 trajectories for $\lambda < 0$, and over 10^6 trajectories for $\lambda \geqslant 0$. There is clear evidence of a transition between a phase of finite and a phase of vanishing entropy production similar to Fig. 1(b) for the driven FA model. Compared to the mean field solution the transition is smeared out and shifted to $\lambda \gtrsim 0$ due to finite size effects. The data suggests that the transition point moves towards $\lambda = 0$ with increasing space-time volume. The probability density of the entropy production rate in Fig. 2(b) for $\lambda = 0.003$ displays a bimodal character, as expected from a first-order transition. Just like for the case of equilibrium dynamics and the space-time transition in terms of activity [8], we expect the first-order transition between phases of distinct entropy production to be generic in driven KCMs.

Mapping ensembles.—We now address the question of whether the biased ensembles of trajectories can be realized experimentally. In the λ -ensemble, the weight of a trajectory segment starting in state \mathcal{C} , surviving for Δt , and then jumping to state \mathcal{C}' reads

$$e^{-r(\mathcal{C})\Delta t}w(\mathcal{C}\to\mathcal{C}')e^{-\lambda\Delta s(\mathcal{C},\mathcal{C}')-N\Delta t\Psi(\lambda)}$$
.

We consider long trajectories and include the large deviation function $\Psi(\lambda)$ for normalization. Collecting terms containing Δt , the exit rate \tilde{r} in a modified dynamics must fulfill

$$\tilde{r}(C;\lambda) = \sum_{C'} \tilde{w}(C \to C';\lambda) = r(C) + N\Psi(\lambda), \quad (6)$$

i.e., the difference between modified and original exit rate is given by $\Psi(\lambda)$ for all states. Simply modifying the transition rates as $we^{-\lambda\Delta s}$ is not enough to give the exit rates \tilde{r} . However, adding a sub-extensive term to $s_{\rm m}$ does not change the large deviation function. We therefore define the modified rates

$$\tilde{w}(\mathcal{C} \to \mathcal{C}'; \lambda) = w(\mathcal{C} \to \mathcal{C}') e^{-\lambda \Delta s(\mathcal{C}, \mathcal{C}') - \Delta u(\mathcal{C}, \mathcal{C}'; \lambda)}$$
(7)

such that the sum of Δu along single trajectories is sub-extensive in $t_{\rm obs}$. Due to antisymmetry $\Delta u(\mathcal{C}',\mathcal{C}) = -\Delta u(\mathcal{C},\mathcal{C}')$ the sum $\sum_{\ell} \Delta u = 0$ vanishes along any closed loop ℓ . This property implies $\Omega - 1$ independent quantities Δu , where Ω is the number of configurations \mathcal{C} . The Ω equations (6) then determine $\{\Psi, \Delta u\}$. A similar result has been found in Ref. [23] for biasing equilibrium towards a shear driven ensemble.

In the simplest case, $\Delta u(\mathcal{C}, \mathcal{C}'; \lambda) \equiv u(\mathcal{C}'; \lambda) - u(\mathcal{C}; \lambda)$ with state function $u(\mathcal{C}; \lambda)$ depending on λ . The sum $\sum_{\alpha=1}^K \Delta u(\mathcal{C}_{\alpha-1}, \mathcal{C}_{\alpha}) = u(\mathcal{C}_K) - u(\mathcal{C}_0)$ is then a temporal boundary term. As we will see below, $u(\mathcal{C})$ can be interpreted as an energy function. The modified dynamics (7) is still Markovian but the rates become nonlocal since a transition from site i can now depend on the whole configuration \mathcal{C} . Even for simple KCMs configuration space is too big to allow an explicit determination of $u(\mathcal{C})$.

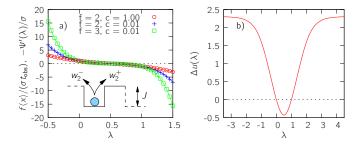


FIG. 3: Particle moving in an infinite lattice with alternating energy levels (see inset). (a) Symbols are numerical results of $f\langle x\rangle/(\sigma t_{\rm obs})$ for a particle moving with rates Eq. (8), where $\sigma \equiv -\Psi'(0)$. The values agree with the normalized mean entropy production rate $-\Psi'(\lambda)/\sigma$ in the λ -ensemble (solid lines). (b) The function $\Delta u(\lambda)$ for f=2 and c=0.01.

To illustrate the idea of mapping the λ -ensemble to a physical dynamics we consider a simple model related to the driven FA model, but without kinetic constraints, that can be tackled analytically. A random walker moves in a periodic lattice with alternating site energies, see inset of Fig. 3(a). The equilibrium rates are $w_1^{\pm}=1$ and $w_2^{\pm}=c$ for steps to the right (+) or left (-), where $c\equiv e^{-\beta J}$ and J is the energy difference. Applying a driving force f, the entropy produced in a single step is $\pm f$. The total entropy production is $s_{\rm m}=fx$, where x is the distance the particle has traveled. The modified rates from Eq. (7) are

$$\tilde{w}_{1}^{+} = e^{+f/2 - \lambda f - \Delta u}, \quad \tilde{w}_{2}^{+} = ce^{+f/2 - \lambda f + \Delta u},$$

$$\tilde{w}_{1}^{-} = e^{-f/2 + \lambda f - \Delta u}, \quad \tilde{w}_{2}^{-} = ce^{-f/2 + \lambda f + \Delta u}$$
(8)

with only one independent Δu . The two equations (6) then lead to quadratic equations for the unknown quantities $\Delta u(\lambda)$ and $\Psi(\lambda)$. The solutions are:

$$\begin{split} e^{\Delta u(\lambda)} &= \frac{1}{2cA(\lambda)} \left[(c-1) + \sqrt{(c-1)^2 + 4c[A(\lambda)]^2} \right], \\ &\Psi(\lambda) = 2c \cosh(f/2) [A(\lambda)e^{\Delta u(\lambda)} - 1] \end{split}$$

with $A(\lambda) \equiv \cosh[f(\lambda - 1/2)]/\cosh(f/2)$. Again, both functions above are symmetric through $A(1 - \lambda) = A(\lambda)$.

Fig. 3(a) shows the mean entropy production in the λ -ensemble of the original driven system with rates $w_{1,2}^{\pm}$. Around $\lambda=0$ and $\lambda=1$ we observe a crossover between regimes of high entropy production and one of low entropy production (something similar has been observed for a driven particle moving in a periodic potential [24]). Fig. 3(a) also shows that the biased entropy production coincides with the typical entropy production in the mapped problem with rates $\tilde{w}_{1,2}^{\pm}$ from Eq. (8). The driving force in the mapped system transforms as $\tilde{f} = f(1-2\lambda)$. The barrier height is adjusted through Δu [see Fig. 3(b)] and the barrier becomes maximal for $\lambda = 1/2$ and vanishes for large $|\lambda|$ with $\Delta u \to \beta J/2$ to allow for the maximal current. One should note that the

correct expression to compare $-\Psi'(\lambda)$ with is $f\langle x\rangle$ [where the brackets are now the average over the dynamics with rates (8)] and not the actual entropy production $\tilde{f}\langle x\rangle$ of the mapped system.

Conclusions.— We have shown that the first-order space-time phase transitions between active and inactive phases observed in the equilibrium dynamics of model glasses [8] are also present in systems driven to non-equilibrium stationary states. In this case the transition is between phases with differing entropy production rates. A next step is to search for evidence of such phase transitions in driven atomistic liquids in analogy with what was done in Ref. [9] for the case of equilibrium fluids.

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